

## From the INTERNATIONAL BUREAU

PCT

NOTIFICATION OF ELECTION  
(PCT Rule 61.2)

Date of mailing (day/month/year) 20 June 2000 (20.06.00)	To:  Assistant Commissioner for Patents United States Patent and Trademark Office Box PCT Washington, D.C.20231 ETATS-UNIS D'AMERIQUE  In its capacity as elected Office
International application No. PCT/US99/24533	Applicant's or agent's file reference 00246/514W03
International filing date (day/month/year) 20 October 1999 (20.10.99)	Priority date (day/month/year) 21 October 1998 (21.10.98)
Applicant GORDON, Roy, G. et al.	

1. The designated Office is hereby notified of its election made:

in the demand filed with the International Preliminary Examining Authority on:

18 May 2000 (18.05.00)

in a notice effecting later election filed with the International Bureau on:

\_\_\_\_\_

2. The election  was

was not

made before the expiration of 19 months from the priority date or, where Rule 32 applies, within the time limit under Rule 32.2(b).

The International Bureau of WIPO 34, chemin des Colombettes 1211 Geneva 20, Switzerland  Facsimile No.: (41-22) 740.14.35	Authorized officer  C. Villet  Telephone No.: (41-22) 338.83.38
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Form PCT/B/331 (July 1992)

US9924533

## PATENT COOPERATION TREATY

PCT

## INTERNATIONAL PRELIMINARY EXAMINATION REPORT

REC'D 13 FEB 2001

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## (PCT Article 36 and Rule 70)

Applicant's or agent's file reference 00246/514WO3	FOR FURTHER ACTION      See Notification of Transmittal of International Preliminary Examination Report (Form PCT/IPEA/416)	
International application No. PCT/US99/24533	International filing date (day/month/year) 20 OCTOBER 1999	Priority date (day/month/year) 21 OCTOBER 1998
International Patent Classification (IPC) or national classification and IPC IPC(7): C23C 16/00, 16/04, 16/18; C07F 3/00 and US Cl.: 427/252; 106/1.05, 1.25; 556/40		
Applicant THE PRESIDENT AND FELLOWS OF HARVARD COLLEGE		

<p>1. This international preliminary examination report has been prepared by this International Preliminary Examining Authority and is transmitted to the applicant according to Article 36.</p> <p>2. This REPORT consists of a total of <u>4</u> sheets.</p> <p><input type="checkbox"/> This report is also accompanied by ANNEXES, i.e., sheets of the description, claims and/or drawings which have been amended and are the basis for this report and/or sheets containing rectifications made before this Authority. (see Rule 70.16 and Section 607 of the Administrative Instructions under the PCT).</p> <p>These annexes consist of a total of <u>0</u> sheets.</p> <p>3. This report contains indications relating to the following items:</p> <ul style="list-style-type: none"> <li>I <input checked="" type="checkbox"/> Basis of the report</li> <li>II <input type="checkbox"/> Priority</li> <li>III <input type="checkbox"/> Non-establishment of report with regard to novelty, inventive step or industrial applicability</li> <li>IV <input type="checkbox"/> Lack of unity of invention</li> <li>V <input checked="" type="checkbox"/> Reasoned statement under Article 35(2) with regard to novelty, inventive step or industrial applicability; citations and explanations supporting such statement</li> <li>VI <input type="checkbox"/> Certain documents cited</li> <li>VII <input type="checkbox"/> Certain defects in the international application</li> <li>VIII <input type="checkbox"/> Certain observations on the international application</li> </ul>
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Date of submission of the demand 18 MAY 2000	Date of completion of this report 28 DECEMBER 2000
Name and mailing address of the IPEA/US Commissioner of Patents and Trademarks Box PCT Washington, D.C. 20231	Authorized officer <i>Dorthea Lawrence</i> JEAN F VOLLAO
Facsimile No. (703) 305-3230	Telephone No. (703) 308-1235

## INTERNATIONAL PRELIMINARY EXAMINATION REPORT

International application No.

PCT/US99/24533

## I. Basis of the report

## 1. With regard to the elements of the international application: \*

 the international application as originally filed the description:pages 1-30 \_\_\_\_\_, as originally filed  
pages NONE \_\_\_\_\_, filed with the demand  
pages NONE \_\_\_\_\_, filed with the letter of \_\_\_\_\_ the claims:pages 31-33 \_\_\_\_\_, as originally filed  
pages NONE \_\_\_\_\_, as amended (together with any statement) under Article 19  
pages NONE \_\_\_\_\_, filed with the demand  
pages NONE \_\_\_\_\_, filed with the letter of \_\_\_\_\_ the drawings:pages 1 \_\_\_\_\_, as originally filed  
pages NONE \_\_\_\_\_, filed with the demand  
pages NONE \_\_\_\_\_, filed with the letter of \_\_\_\_\_ the sequence listing part of the description:pages NONE \_\_\_\_\_, as originally filed  
pages NONE \_\_\_\_\_, filed with the demand  
pages NONE \_\_\_\_\_, filed with the letter of \_\_\_\_\_

## 2. With regard to the language, all the elements marked above were available or furnished to this Authority in the language in which the international application was filed, unless otherwise indicated under this item.

These elements were available or furnished to this Authority in the following language \_\_\_\_\_ which is:

the language of a translation furnished for the purposes of international search (under Rule 23.1(b)).  
 the language of publication of the international application (under Rule 48.3(b)).  
 the language of the translation furnished for the purposes of international preliminary examination (under Rules 55.2 and/or 55.3).

## 3. With regard to any nucleotide and/or amino acid sequence disclosed in the international application, the international preliminary examination was carried out on the basis of the sequence listing:

contained in the international application in printed form.  
 filed together with the international application in computer readable form.  
 furnished subsequently to this Authority in written form.  
 furnished subsequently to this Authority in computer readable form.  
 The statement that the subsequently furnished written sequence listing does not go beyond the disclosure in the international application as filed has been furnished.  
 The statement that the information recorded in computer readable form is identical to the written sequence listing has been furnished.

4.  The amendments have resulted in the cancellation of:

the description, pages NONE  
 the claims, Nos. NONE  
 the drawings, sheets/fig NONE

5.  This report has been drawn as if (some of) the amendments had not been made, since they have been considered to go beyond the disclosure as filed, as indicated in the Supplemental Box (Rule 70.2(c)).\*\*

\* Replacement sheets which have been furnished to the receiving Office in response to an invitation under Article 14 are referred to in this report as "originally filed" and are not annexed to this report since they do not contain amendments (Rules 70.16 and 70.17).

\*\*Any replacement sheet containing such amendments must be referred to under item 1 and annexed to this report.

## INTERNATIONAL PRELIMINARY EXAMINATION REPORT

International application No.

PCT/US99/24533

**V. Reasoned statement under Article 35(2) with regard to novelty, inventive step or industrial applicability; citations and explanations supporting such statement****1. statement**

Novelty (N)	Claims <u>10-13</u>	YES
	Claims <u>1-9, 14-19 and 21</u>	NO
Inventive Step (IS)	Claims <u>10-13</u>	YES
	Claims <u>1-9, 14-21</u>	NO
Industrial Applicability (IA)	Claims <u>1-21</u>	YES
	Claims <u>NONE</u>	NO

**2. citations and explanations (Rule 70.7)**

Claims 1-9, 14-19 and 21 lack novelty under PCT Article 33(2) as being anticipated by Gordon et al (US 5139999).

Gordon et al teaches chemical vapor deposition processes using alkaline earth metal precursors including beta diketonates ( column 4, lines 4-39).

Gordon et al teaches that the precursor is in combination with an amine moiety including tetramethylethylenediamine and other amine multidentates.

Gordon et al teaches that the compounds include barium dipivaloylmethane and barium hexafluoroacetylacetone.

Gorden et al teaches that the metals can be selected from strontium and barium.

When Gordon et al teaches that the deposited material can be one or more alkaline earth metal oxides (column 5, lines 37-50)and can be deposited by various methods including spin coating then the claims are anticipated(column 5, line 27).

Claims 1, 16 and 20 lack an inventive step under PCT Article 33(3) as being obvious over Gordon et al (US 5139999).

Gordon et al teaches chemical vapor deposition processes using alkaline earth metal precursors including beta diketonates ( column 4, lines 4-39).

Gorden et al teaches that the alkaline eart metal beta metal diketonate and amine are heated to a vapor and deposit a material containing alkaline metal oxides on a substrate ( column 5).

Gorden et al does not specifically teach a sol-gel process. However the sol-gel process is a common process in the art for deposition of an alkaline earth metal material and Gorden et al teaches that any commonly known process can be used for the deposition. It would have been obvious to one of ordinary skill in the art to have used the process of Gorden et al and to have used the commonly known sol-gel process for the deposition.

Claim 1-3,5, and 7-8 lack novelty under PCT Article 33(2) as being anticipated by rimmer et al (US5248787).

When Trimmer et al teaches volatile barium, strontium and calcium beta diketonates with a nitrogen donor that includes tetramethyl ethylenediamine ( see abstract, column 1, lines 40-68 (Continued on Supplemental Sheet.)

**Supplemental Box**

(To be used when the space in any of the preceding boxes is not sufficient)

Continuation of: Boxes I - VIII

Sheet 10

**V. 2. REASoNED STATEMENTS - CITATIONS AND EXPLANATIONS (Continued):**  
and column 2, lines 49-50) then the claims are anticipated.

Claims 1-3,5, 7-8, 14-19 lack inventive step under PCT Article 33(3) as being obvious over Trimmer et al (USS248787).

Trimmer et al teaches volatile barium, strontium and calcium beta diketonates with a nitrogen donor that includes tetramethyl ethylenediamine ( see abstract, column 1, lines 40-68 and column 2, lines 49-50).

Trimmer et al teaches that the compounds are deposited on a substrate by spin coating with MO-CVD techniques ( column 4, lines 1-25) to form oxides such as calcium, strontium or barium oxides.

Although the use of amine composition of beta diketonates of alkaline metals is not exemplified it is expressly suggested.

Claims 1-4 and 6 lack novelty under PCT Article 33(2) as being anticipated by Chem abs 359 ( CA:118:70359).

When Chem abs 359 teaches beta diketonate compounds of barium with the triethyl amine wherein the beta diketonate is ,2,6,6-tetramethyl 3,5-heptanedione then the claims are anticipated.

Claims 10-13 the criteria set out in PCT Article 33(2)-(4), because the prior art does not teach or fairly suggest compounds and amines of the formula/composition found in instant claims 10-13.

## ----- NEW CITATIONS -----

NONE

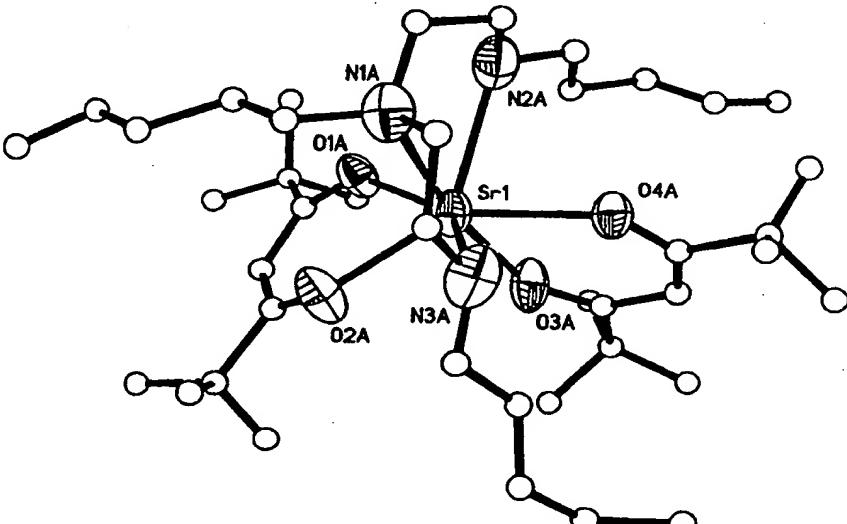


## INTERNATIONAL APPLICATION PUBLISHED UNDER THE PATENT COOPERATION TREATY (PCT)

(51) International Patent Classification <sup>7</sup> : <b>C23C 16/00, 16/04, 16/18, C07F 3/00</b>		A1	(11) International Publication Number: <b>WO 00/23635</b>
			(43) International Publication Date: <b>27 April 2000 (27.04.00)</b>
(21) International Application Number:	PCT/US99/24533		
(22) International Filing Date:	20 October 1999 (20.10.99)		
(30) Priority Data:	60/105,158 60/126,793	21 October 1998 (21.10.98) 30 March 1999 (30.03.99)	US US
(71) Applicant (for all designated States except US):	THE PRESIDENT AND FELLOWS OF HARVARD COLLEGE [US/US]; 17 Quincy Street, Cambridge, MA 02138 (US).		
(72) Inventors; and			
(73) Inventors/Applicants (for US only):	GORDON, Roy, G. [US/US]; 22 Highland Street, Cambridge, MA 02138 (US). TEFF, Daniel [US/US]; Apartment 2007, Unit 4/28-Building 3, 7125 East Superstition Springs Boulevard, Mesa, AZ 85208 (US).		
(74) Agent:	SCOZZAFAVA, Mary, Rose; Clark & Elbing LLP, 176 Federal Street, Boston, MA 02110-2214 (US).		

**(54) Title:** LIQUID COMPOUNDS FOR FORMATION OF MATERIALS CONTAINING ALKALINE EARTH METALS**(57) Abstract**

A liquid precursor is provided for the formation of alkaline earth containing materials. The liquid precursor comprises an alkaline earth metal beta-diketonate bound to an amine. For example, a liquid compound was formed by reacting N,N',N''-trihexyldiethylenetriamine with barium 2,2,6,6-tetramethyl-3,5 heptanedionate. Films containing alkaline earth metals are deposited from vapors of the precursor liquids and, optionally, oxygen or other sources of oxygen. This process may be used to deposit barium strontium titanate films having a high dielectric constant. The liquid precursors may also be used for spray coating and sol-gel deposition of materials. The figure is an X-ray crystallographic structure of strontium bis (2,2,6,6-tetramethyl-heptane-3,5-dionate) with N,N',N''-triamyldiethylenetriamine.



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DK	Denmark	LR	Liberia	SG	Singapore		
EE	Estonia						

## INTERNATIONAL SEARCH REPORT

International application No.

PCT/US99/24533

## A. CLASSIFICATION OF SUBJECT MATTER

IPC(7) :C23C 16/00, 16/04, 16/18; C07F 3/00  
US CL :427/252; 106/1.05, 1.25; 556/40

According to International Patent Classification (IPC) or to both national classification and IPC

## B. FIELDS SEARCHED

Minimum documentation searched (classification system followed by classification symbols)

U.S. : 427/252; 106/1.05, 1.25

Documentation searched other than minimum documentation to the extent that such documents are included in the fields searched

Electronic data base consulted during the international search (name of data base and, where practicable, search terms used)

Please See Extra Sheet.

## C. DOCUMENTS CONSIDERED TO BE RELEVANT

Category*	Citation of document, with indication, where appropriate, of the relevant passages	Relevant to claim No.
Y	US 5,248,787 A (Timmer et al) 28 September 1993 (28.09.93), see column 2, lines 42-57.	1-5, 7-8, 14-21
A	US 5,280,012 A (KIRLIN et al) 18 January 1994 (18.01.94), see columns 1-6.	1-21
A	US 5,225,561 A (KIRLIN et al) 06 July 1993 (06.07.93), see columns 4-6.	1-21
Y	US 5,139,999 A (GORDON et al) 18 August 1992 (18.08.92), see columns 4-5.	1-21

Further documents are listed in the continuation of Box C.  See patent family annex.

* Special categories of cited documents:	"T"	later document published after the international filing date or priority date and not in conflict with the application but cited to understand the principle or theory underlying the invention
"A" document defining the general state of the art which is not considered to be of particular relevance	"X"	document of particular relevance; the claimed invention cannot be considered novel or cannot be considered to involve an inventive step when the document is taken alone
"E" earlier document published on or after the international filing date	"Y"	document of particular relevance; the claimed invention cannot be considered to involve an inventive step when the document is combined with one or more other such documents, such combination being obvious to a person skilled in the art
"L" document which may throw doubts on priority claim(s) or which is cited to establish the publication date of another citation or other special reason (as specified)	"&"	document member of the same patent family
"O" document referring to an oral disclosure, use, exhibition or other means		
"P" document published prior to the international filing date but later than the priority date claimed		

Date of the actual completion of the international search	Date of mailing of the international search report
26 DECEMBER 1999	08 MAR 2000
Name and mailing address of the ISA/US Commissioner of Patents and Trademarks Box PCT Washington, D.C. 20231 Facsimile No. (703) 305-3230	Authorized officer JEAN F VOLLANO Telephone No. (703) 308-1235

INTERNATIONAL SEARCH REPORT

International application No.

PCT/US99/24533

B. FIELDS SEARCHED

Electronic data bases consulted (Name of data base and where practicable terms used):

EAST, CAS ONLINE, GMELIN

search: dionate, barium, calcium, magnesium, strontium, gel-sol, spin-coating, spray-coating, amine, diamine, triamine, cvd, chemical vapor deposition

## INTERNATIONAL SEARCH REPORT

International Application No

PCT/US 98/07829

**A. CLASSIFICATION OF SUBJECT MATTER**  
 IPC 6 C07F19/00 C23C16/00

According to International Patent Classification (IPC) or to both national classification and IPC

**B. FIELDS SEARCHED**Minimum documentation searched (classification system followed by classification symbols)  
 IPC 6 C07F C23C

Documentation searched other than minimum documentation to the extent that such documents are included in the fields searched

Electronic data base consulted during the international search (name of data base and, where practical, search terms used)

**C. DOCUMENTS CONSIDERED TO BE RELEVANT**

Category	Citation of document, with indication, where appropriate, of the relevant passages	Relevant to claim No.
A	<p>CHEMICAL ABSTRACTS, vol. 103, no. 12,    23 September 1985    Columbus, Ohio, US;    abstract no. 93770,    MALHOTRA, R. K. ET AL: "Synergism in the    extraction of uranium(VI) by a mixture of    oxine and beta.- diketones"    XP002070941    see abstract    &amp; PROC. - INDIAN ACAD. SCI., CHEM. SCI.    (1985), 94(3), 515-19 CODEN: PIAADM; ISSN:    0253-4134,    1985,</p> <p>---</p> <p style="text-align: center;">-/-</p>	1

 Further documents are listed in the continuation of box C. Patent family members are listed in annex.

## \* Special categories of cited documents :

- "A" document defining the general state of the art which is not considered to be of particular relevance
- "E" earlier document but published on or after the international filing date
- "L" document which may throw doubts on priority claim(s) or which is cited to establish the publication date of another citation or other special reason (as specified)
- "O" document referring to an oral disclosure, use, exhibition or other means
- "P" document published prior to the international filing date but later than the priority date claimed

"T" later document published after the international filing date or priority date and not in conflict with the application but cited to understand the principle or theory underlying the invention

"X" document of particular relevance; the claimed Invention cannot be considered novel or cannot be considered to involve an inventive step when the document is taken alone

"Y" document of particular relevance; the claimed Invention cannot be considered to involve an inventive step when the document is combined with one or more other such documents, such combination being obvious to a person skilled in the art.

"&" document member of the same patent family

Date of the actual completion of the international search

Date of mailing of the International search report

9 July 1998

20/08/1998

Name and mailing address of the ISA  
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Authorized officer

Rinkel, L

## INTERNATIONAL SEARCH REPORT

International Application No
PCT/US 98/07829

## C.(Continuation) DOCUMENTS CONSIDERED TO BE RELEVANT

Category	Citation of document, with indication, where appropriate, of the relevant passages	Relevant to claim No.
A	<p>CHEMICAL ABSTRACTS, vol. 71, no. 8,  25 August 1969  Columbus, Ohio, US;  abstract no. 33898,  WOO, CHING-HUNG: "Extractions of several  rare earths using a combination of  thenoyltrifluoroacetone and acetylacetone"  XP002070942  see abstract  &amp; U. S. AT. ENERGY COMM. (1968),  ORO-2124-18, 94 PP. AVAIL.: DEP.;CFSTI  FROM: NUCL. SCI. ABSTR. 1969, 23(7), 11605  CODEN: XAERAK,  1968,</p> <p>EP 0 527 661 A (THE ASSOCIATED OCTEL  COMPANY LIMITED) 17 February 1993  see the whole document</p>	1
A		1-24

# INTERNATIONAL SEARCH REPORT

## Information on patent family members

Int'l. Application No

PCT/US 98/07829

Patent document cited in search report	Publication date	Patent family member(s)		Publication date
EP 527661	A 17-02-1993	AU	658922 B	04-05-1995
		AU	2437692 A	16-03-1993
		CA	2113715 A	04-03-1993
		WO	9304072 A	04-03-1993
		JP	7500318 T	12-01-1995
		NO	940471 A	13-04-1994